

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US00/29822

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : H01J 1/62, 63/04

US CL : 313/495, 496, 497, 498, 499, 500, 501, 502, 503, 504, 505, 506, 507, 508, 509, 512; 428/690, 917; 445/24

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 313/495, 496, 497, 498, 499, 500, 501, 502, 503, 504, 505, 506, 507, 508, 509, 512; 428/690, 917; 445/24

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EAST

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5,371,434 A (RAWLINGS) 06 December 1994 (06.12.1994)	1-20
A	US 5,618,626 A (NAGASHIMA et al.) 08 April 1997 (08.04.1997)	1-20
A	US 5,814,416 A (DODABALAPUR et al.) 29 September 1998 (29.09.1998)	1-20
A	US 5,936,347 A (ISAKA et al.) 10 August 1999 (10.08.1999)	1-20

☐ Further documents are listed in the continuation of Box C.

☐ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T"

later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X"

document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y"

document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents; such combination being obvious to a person skilled in the art

"&"

document member of the same patent family

Date of the actual completion of the international search

16 January 2001 (16.01.2001)

Date of mailing of the international search report

23 FEB 2001

Name and mailing address of the ISA/US

Commissioner of Patents and Trademarks
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PCT COOPERATION TREATY

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Commissioner
 US Department of Commerce
 United States Patent and Trademark
 Office, PCT
 2011 South Clark Place Room
 CP2/5C24
 Arlington, VA 22202
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 in its capacity as elected Office

Date of mailing (day/month/year) 24 July 2001 (24.07.01)	
International application No. PCT/US00/29822	Applicant's or agent's file reference 7616/42
International filing date (day/month/year) 30 October 2000 (30.10.00)	Priority date (day/month/year) 29 October 1999 (29.10.99)
Applicant STURM, James, C. et al	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:
 14 May 2001 (14.05.01)

☐ in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was
☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35	Authorized officer Charlotte ENGER Telephone No.: (41-22) 338.83.38
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10/08983m

PATENT COOPERATION TREATY

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REC'D 12 JUN 2002

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INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

8

Applicant's or agent's file reference 7616/42	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/US00/29822	International filing date (day/month/year) 30 October 2000 (30.10.2000)	Priority date (day/month/year) 29 October 1999 (29.10.1999)
International Patent Classification (IPC) or national classification and IPC IPC(7): H01J 1/62, 63/04; H05B 33/00, 33/02, 33/26 and US Cl.: 313/495, 496, 497, 498, 499, 500, 501, 502, 503, 504, 505, 506, 507, 508, 509, 512; 428/690, 917; 445/24		
Applicant TRUSTEES OF PRINCETON UNIVERSITY		

- This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
- This REPORT consists of a total of 6 sheets, including this cover sheet.
☐ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 0 sheets.

- This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of report with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand 14 May 2001 (14.05.2001)	Date of completion of this report 17 May 2002 (17.05.2002)
Name and mailing address of the IPEA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703)305-3230	Authorized officer Nimeshkumar D Patel <i>Nimeshkumar D Patel</i> Telephone No. (703)308-0956

Form PCT/IPEA/409 (cover sheet)(July 1998)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.

PCT/US00/29822

I. Basis of the report

1. With regard to the elements of the international application:*

- ☒ the international application as originally filed.
- ☒ the description:
pages 1-12 _____ as originally filed
pages NONE _____, filed with the demand
pages NONE _____, filed with the letter of _____.
- ☒ the claims:
pages 13-15 _____, as originally filed
pages NONE _____, as amended (together with any statement) under Article 19
pages NONE _____, filed with the demand
pages NONE _____, filed with the letter of _____.
- ☒ the drawings:
pages 1-3 _____, as originally filed
pages NONE _____, filed with the demand
pages NONE _____, filed with the letter of _____.
- ☐ the sequence listing part of the description:
pages NONE _____, as originally filed
pages NONE _____, filed with the demand
pages NONE _____, filed with the letter of _____.

2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language _____ which is:

- ☐ the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of the translation furnished for the purposes of international preliminary examination (under Rules 55.2 and/or 55.3).

3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in printed form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. ☐ The amendments have resulted in the cancellation of:

- ☐ the description, pages NONE
- ☐ the claims, Nos. NONE
- ☐ the drawings, sheets/~~fig~~ NONE

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).**

* Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17).

** Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.
PCT/US00/29822

V. Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. STATEMENT

Novelty (N)	Claims <u>NONE</u>	YES
	Claims <u>1-20</u>	NO
Inventive Step (IS)	Claims <u>NONE</u>	YES
	Claims <u>1-20</u>	NO
Industrial Applicability (IA)	Claims <u>1-20</u>	YES
	Claims <u>NONE</u>	NO

2. CITATIONS AND EXPLANATIONS

Please See Continuation Sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the questions whether the claims are fully supported by the description, are made:

Claim 1 is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because claim 1 is indefinite for the following reason(s):

In line 5, claim 1 recites the limitation "said conductive oxide layer". There is insufficient antecedent basis for this limitation.

Claim 4 is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because claim 1 is indefinite for the following reason(s):

In lines 2-3, claim 4 recites the limitation "a second transparent layer". There is insufficient antecedent basis for this limitation.

Claim 8 is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because claim 1 is indefinite for the following reason(s):

In line 3, claim 8 recites the limitation "a second transparent layer". There is insufficient antecedent basis for this limitation.

Claim 12 is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because claim 1 is indefinite for the following reason(s):

In line 3, claim 12 recites the limitation "a second transparent layer". There is insufficient antecedent basis for this limitation.

Claim 15 is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because claim 1 is indefinite for the following reason(s):

In line 5, claim 15 recites the limitation "a second transparent layer". There is insufficient antecedent basis for this limitation.

Claim 20 is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because claim 1 is indefinite for the following reason(s):

In line 3, claim 20 recites the limitation "a second transparent layer". There is insufficient antecedent basis for this limitation.

Claims 1, 3-5, 7-9, 11, 12, 15, 17, 19 and 20 is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because claim 1 is indefinite for the following reason(s):

Claims 1, 3-5, 7-9, 11, 12, 15, 17, 19 and 20 recite the limitation "assume a non-planar form", the term "assume" renders the claim indefinite since the meaning of the word does not clearly define the structure. The word assume means to pretend to be or form, consequently, is considered indefinite because it does not clearly define the feature claimed.

Supplemental Box

(To be used when the space in any of the preceding boxes is not sufficient)

Regarding claim 9, Isaka discloses a method for constructing a light emitting device comprising the steps of providing a transparent substrate (9) having a first (top of substrate 9) and a second surface (bottom of substrate 9) of the substrate, depositing an OLED layer (12) on the conductive transparent layer (10), and causing a contour of the second surface (bottom of substrate 9) of the substrate to assume a non-planar form (Column 8, lines 8-21).

Regarding claim 10, Isaka discloses a method for constructing a light-emitting device wherein the non-planar form is spherical (Fig. 5, element 41).

Regarding claim 11, Isaka discloses a method for constructing a light emitting device wherein the second surface (bottom of substrate 9) contour is caused to assume a non-planar form by moulding the substrate surface (Column 8, lines 8-21).

Regarding claim 12, Isaka discloses a method for constructing a light emitting device wherein the second surface (bottom of substrate 9) contour is caused to assume a non-planar form laminating a first surface of a second transparent layer having a non-planar second surface to the second surface of the substrate (Column 8, lines 8-21).

Regarding claim 13, Isaka discloses a light emitting device comprising a transparent substrate (9) having a first (top of substrate 9) and second surface (bottom of substrate 9), an OLED layer (12) disposed on the first surface (top of substrate 9) of the substrate, wherein the second surface (bottom of substrate 9) of the substrate is molded to a non-planar form (Column 8, lines 8-21).

Regarding claim 14, Isaka discloses a light-emitting device wherein the non-planar form is spherical (Fig. 5, element 41).

Regarding claim 15, Isaka discloses a light emitting device comprising a transparent substrate (9) having a first (top of substrate 9) and second surface (bottom of substrate 9), an OLED layer (12) disposed on the first surface (top of substrate 9) of the substrate, wherein the second surface (bottom of substrate 9) of the substrate is caused to assume a non-planar form by laminating a first surface of a second transparent layer having a non-planar second surface to the second surface of the substrate (Column 8, lines 8-21).

Regarding claim 16, Isaka discloses a light-emitting device wherein the non-planar form is spherical (Fig. 5, element 41).

Regarding claim 17, Isaka discloses a method for increasing light emissivity of a light emitting device, wherein the light emitting device includes at least two OLEDs (12) disposed on a first surface of a transparent substrate (9), the method comprising the step of causing a contour of selected portions of a second surface of the substrate to assume a non-planar form, the selected portions being selected to be optical alignment with the light output from ones of the at least two OLEDs (Column 8, lines 8-21).

Regarding claim 18, Isaka discloses a method for increasing light emissivity of a light emitting device wherein the non-planar form is spherical (Fig. 5, element 41).

Regarding claim 19, Isaka discloses a method for increasing light emissivity of a light emitting device wherein the selected second surface contours are caused to assume a non-planar form by moulding the substrate surface (Column 8, lines 8-21).

Regarding claim 20, Isaka discloses a method for increasing light emissivity of a light emitting device wherein the selected second surface contours are caused to assume a non-planar form by laminating a first surface of a second transparent layer having a non-planar second surface to the second surface of the substrate (Column 8, lines 8-21).

Claims 1-20 meet the criteria set out in PCT Article 33(4), because they can be manufactured and are deemed useful in industry.



Supplemental Box

(To be used when the space in any of the preceding boxes is not sufficient)

V. 2. Citations and Explanations:

Claims 1-20 lack novelty under PCT Article 33(2) as being anticipated by Isaka et al. (US 5,936,347).

Regarding claim 1, Isaka discloses a light emitting device (see Fig.5) comprising a transparent substrate (9) having a first surface (top of substrate 9) and a second surface (bottom of substrate 9), a conductive transparent layer (10) disposed on the first surface of the substrate (9), an organic layer (41) disposed on the conductive layer (10), and a top contact (6) disposed on the organic layer, wherein a contour of the second surface (bottom of substrate 9) of the substrate is caused to assume a non-planar form (41 in Figure 5, concave and convex surface, Column 8, lines 8-22) .

Regarding claim 2, Isaka discloses a light-emitting device wherein the non-planar form is spherical (Fig. 5, element 41).

Regarding claim 3, Isaka discloses a light emitting device wherein the second surface contour is caused to assume a non-planar form by moulding the substrate surface. The Examiner notes that the claim limitation that " is caused to assume a non-planar form by moulding the substrate surface" is drawn to a process of manufacturing which is incidental to the claimed apparatus. It is well established that a claimed apparatus can not be distinguished over the prior art by a process limitation. Consequently, absent a showing of an unobvious difference between the claimed product and the prior art, the subject product-by-process claim limitation is not afforded patentable weight.

Regarding claim 4, Isaka discloses a light emitting device wherein the second surface (bottom of substrate 9) contour is caused to assume a non-planar form by laminating a first surface of a second transparent layer having a non-planar second surface to the second surface of the substrate (Column 8, lines 8-21).

Regarding claim 5, Isaka discloses a method for increasing light emissivity for organic light emitting diodes (OLED), wherein the OLED is disposed on a first surface of a transparent substrate, the method comprising the step of causing a second surface of the substrate to assume a non-planar form (Column 8, lines 8-21).

Regarding claim 6, Isaka discloses a method for increasing light emissivity for OLEDs wherein the non-planar form is spherical (Fig. 5, element 41).

Regarding claim 7, Isaka discloses a method for increasing light emissivity for OLEDs wherein the second surface (bottom of substrate 9) contour is caused to assume a non-planar form by moulding the substrate surface (Column 8, lines 8-21).

Regarding claim 8, Isaka discloses a method for increasing light emissivity for OLEDs wherein the second surface (bottom of substrate 9) contour is caused to assume a non-planar form by laminating a first surface of a second transparent layer having a non-planar second surface to the second surface of the substrate (Column 8, lines 8-21).



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60/162,552 29 October 1999 (29.10.1999) US

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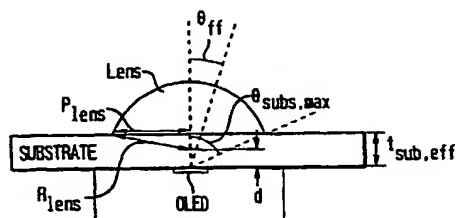
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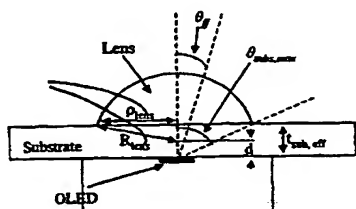
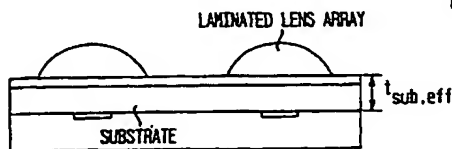
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[Continued on next page]

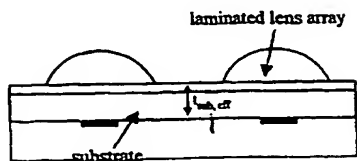
(54) Title: ORGANIC LIGHT EMITTING DIODE HAVING SPHERICAL SHAPED PATTERNS



B



(a)



(b)

(57) Abstract: A light emitting device having a transparent substrate with a first surface and a second surface, a conductive transparent layer on the first surface, an organic layer disposed on the conductive transparent layer, an organic layer disposed on the conductive transparent layer and a top contact disposed on the organic layer, the contour of the second surface of the substrate having a non-planar form.

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Published:

— *With international search report.*

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

ORGANIC LIGHT EMITTING DIODE HAVING SPHERICAL SHAPED PATTERNS

SPECIFICATIONBACKGROUND OF THE INVENTIONRELATED APPLICATIONS

The present invention is related to and claims the benefit of U.S. Provisional Patent Application Serial No. 60/162,552, filed October 29, 1999 and
5 entitled "Improvement of Output Coupling Efficiency of Organic Light Emitting Diodes by Backside Substrate Patterning", which provisional application is assigned to the same assignee and is incorporated by reference herein.

FIELD OF THE INVENTION

10 This invention relates to the field of light emitting devices, and more particularly to organic light emitting devices (OLEDs) and the emission efficiency thereof.

PRIOR ART

15 Access to the Internet and the need to download and view vast quantities of data at greater and greater speeds, along with a frequent requirement for portability and a small footprint are placing greater demands on the capabilities of display devices. The display device of choice for such applications is a flat-panel display, but the current liquid crystal display (LCD) technology in use by most
20 flat-panel displays is limited in its ability to meet these increasing demands. A new display technology, however, offers considerable promise for overcoming the limitations of the LCD technology. That new technology is based on the application of organic light-emitting diodes (OLEDs), which make use of thin film materials that emit light when excited by an electric current.

25 The typical OLED consists of a multi-layer sandwich of a planar glass substrate ($t_{sub} \sim 1\text{mm}$, $n_{sub} = 1.51$), a layer of Indium Tin Oxide (ITO) ($t_{ITO} \sim 100\text{ nm}$, $n_{ITO} \sim 1.8$), one or more organic layers ($t_{org} \sim 0.1\text{ nm}$, $n_{org} = 1.6 - 1.8$), and a reflecting cathode (e.g. Mg:Ag or Li:Al), where t refers to the layer thickness and

n refers to the layer index of refraction. For simplicity, the discussion herein will be based on a single organic layer, where the light emission occurs. However, those skilled in the art will readily understand that the discussion and analysis that follows can readily be extended to more complicated device structures.

5 An important figure of merit for a display system is the efficiency of conversion of input power to emitted light. In OLED displays, a critical factor in determining this system efficiency is the coupling efficiency (η_{ext}) with which internally generated light is coupled out of the device. In order to meet expected demands of future display systems, there is a need to improve the coupling
10 efficiency of OLEDs.

OBJECTS AND SUMMARY OF THE INVENTION

It is accordingly an object of the invention to provide an increase in coupling efficiency for OLEDs. To that end, an approach is provided for increasing the emission intensity of an organic light emitting diode (OLED) and the total external emission efficiency for an OLED at a normal viewing angle. With the approach of the invention, they have been increased by factors of 9.6 and 3.0 respectively by applying spherically shaped patterns to the back of the device substrate. The inventive approach captures light previously lost to wave-guiding in the substrate and, with proper choice of substrate, light previously lost to wave-guiding in the organic/anode layers. According to the method of the invention, a surface texturing approach is provided which, when compared with devices fabricated on typical planar glass substrates, can at least double the emission efficiency for an OLED when glass substrates are used, and at least triple the OLED emission efficiency when high index plastic substrates are used.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 shows ray diagrams for various light paths in a planar OLED.

Figures 2(a) and 2(b) provide a schematic depiction of the use of spherical surface features to improve light emission efficiency for an OLED according to the invention.

Figure 3(a) shows measured far field intensity distribution pattern for planar glass substrate and expected profiles of a Lambertian emitter.

Figure 3(b) shows experimental results for glass substrate devices with and without spherical surface features applied according to the invention.

Figure 3(c) shows experimental results for PC substrate devices with and without spherical surface features applied according to the invention, along with the planar glass substrate results.

DETAILED DESCRIPTION OF THE INVENTION

A novel approach is described herein for significantly improving the emission efficiency of an OLED. In the description of that approach, an analytical construct is first developed to provide an evaluation tool useful for describing the emission efficiency improvement of the invention and comparing that result with prior-art techniques. With that analytical construct in place, the approach of the invention for achieving the improved OLED emission efficiency is then described. Finally, a number of embodiments for the improved emission-efficiency approach of the invention have been implemented by the inventors, and those embodiments are described.

As described in the background section, the coupling efficiency (η_{ext}) of an OLED is a critical factor in the determination of emission efficiency for an OLED display. It is straightforward to analyze η_{ext} for the type of layered structure used in an OLED by considering the indices of refraction associated with each layer. As a predicate to that analysis, consider the ray diagram for planar OLEDs shown in Figure 1, demonstrating loss by light trapping in the substrate layer (ray II) and in the organic/anode layers (ray III). As shown by ray I of the figure, only light emitted at sufficiently small angles will escape.

Because the refraction index for the substrate is less than the organic layer refraction index (*i.e.*, $n_{subs} < n_{org}$), a critical angle, $\theta_{org, c2}$, can be obtained, defined by $\sin^{-1}(n_{subs}/n_{org})$, for which light emitted in the organic layer at an angle greater than $\theta_{org, c2}$ is wave-guided within the ITO and organic layers. This light emission path is illustrated as ray III in Figure 1. Similarly, because $n_{glass} < n_{subs}$, one can also obtain a critical angle, $\theta_{org, c1}$, defined by $\sin^{-1}(n_{air}/n_{org})$, for which light emitted in the organic layer at an angle greater than $\theta_{org, c1}$ is wave-guided in the substrate -- illustrated as ray II in Figure 1. Since only the light emitting at angles less $\theta_{org, c2}$ is emitted from the device -- illustrated as ray I in Figure 1, all of the remaining wave-guided light is effectively lost, representing a reduction in η_{ext} . Applying ray optics and assuming isotropic emission from a point source in the organic layer and a transmission coefficient, T , of 1 for $\theta < \theta_{org, c1}$ and 0 otherwise, one can compute η_{ext} and η_{subs} , the latter representing the fraction of emitted light which is waveguided in the substrate

$$\eta_{ext} = \int_0^{\theta_{org.cl}} \sin \theta d\theta = 1 - \cos \theta_{org.cl} \approx \frac{1}{2n_{org}^2} \text{ (planar substrate), (1)}$$

$$\eta_{subs} = \cos \theta_{org.cl} - \cos \theta_{org.c2}. \quad (2)$$

[See, N. C. Greenham, R. H. Friend, and D. D. C. Bradley, "Angular dependence of the emission from a conjugated polymer light-emitting diode: implications for efficiency calculations", Adv. Mat. 6, 491 (1994)]

Furthermore, the external luminous intensity distribution, under the same assumptions, is given by

$$I_{ext}(\theta_{ff}) = \frac{F}{2\pi} \frac{n_{air}^2 \cos \theta_{ff}}{n_{org}^2 \sqrt{1 - \left(\frac{n_{air}}{n_{org}} \sin \theta_{ff} \right)^2}} \text{ (planar substrate).} \quad (3)$$

[See, G. Gu, D. Z. Garbuzov, P. E. Burrows, S. Venkatesh, and S. R. Forrest, "High-external-quantum-efficiency organic light-emitting devices", Opt. Lett. 22, 396 (1997)]

which resembles the cosine intensity profile of a Lambertian emitter. (Note that the above equations and modeling all ignore well-known microcavity effects that complicate the model but do not change the qualitative efficacy of the methodology and results described herein.)

It should be noted that the assumption that $T=1$ for $\theta < \theta_{org.cl}$ represents a simplification. In particular, it represents the upper bound on the expected emitted intensity. The lower bound (neglecting microcavity effects) can be obtained if one uses the expressions for $T(\theta)$ determined by applying the Fresnel Equations at each interface. However, the simplification can be seen to provide a close approximation. In the determination of the factors represented by equations (1), (2), and (3) for the case of $T=1$ for $\theta < \theta_{org.cl}$, it is implicitly assumed that all of the light internally reflected at these angles is eventually emitted, while in the second approach (lower bound) it is assumed that *none* of the internally reflected light is readmitted. The results obtained for $I_{ext}(\theta_{ff})$ in both cases, along with the cosine result obtained for a Lambertian emitter, are plotted in Figure 3a, along side the experimental results obtained for an OLED fabricated on a planar, glass

substrate. The difference between the two refraction models (*i.e.* $R=1$, representing the case in all reflected light eventually escapes, and $R=0$, representing the case in which all reflected light is lost) is small and it is therefore generally valid to employ either assumption. Since the expressions obtained
 5 under the $T=1$ assumption are simpler, they will be used in for the remainder of this description.

For the expected range of the index of refraction for the organic layer -- between 1.6 and 1.8, it can be seen from Equation (1), that the corresponding range for the coupling efficiency, η_{ext} , will be between 0.20 and 0.15,
 10 demonstrating the significance of the coupling efficiency in degrading system efficiency -- *i.e.* between 80 and 85 percent of the internally generated light is trapped within the device. The external coupling efficiency has been improved by a factor of 1.9 ± 0.2 by etching grooves in the glass around the OLED to redirect light trapped in the substrate and organic/ITO layers [*See, G. Gu, et al., id.*] This
 15 method does not lend itself well to the fabrication of device arrays, however, where metal lines and/or circuitry for passive or active matrix drivers would have to cross the deep grooves. This method also has the disadvantage of requiring the etching of finely controlled, non-vertical profiles in the substrate, significantly increasing the complexity of fabrication.

20 According to the method of the invention, a solution to the light-trapping problem is provided by patterning the back (viewing) side of the substrate in the shape of a sphere with its center at the source of light. Most of the light would then impinge normally on the air-substrate interface, sharply reducing the loss of light due to wave-guiding in the substrate. Such spherical patterning of the
 25 substrate is illustrated schematically in Figure 2.

With reference to the figure, the attachment of a sphere to the backside of the substrate, or shaping the substrate into such a spherical form, shown in Figure 2(a) permits the light rays to escape the substrate at much greater angles. The maximum angle of the light ray that is emitted externally is increased from $\theta_{org, cl}$
 30 in the planar device to $\theta_{max} = \tan^{-1}(\rho_{lens}/t_{subs})$ in the backside-patterned device of the invention (illustrated in Figure 2(a)). To the extent that θ_{max} is determined by the actual pattern and can be made large, the external coupling efficiency is

increased to $\eta_{ext} = 1 - \cos \theta_{max}$ (arrived at by changing the upper limit of integration in Equation (1)). In the center of the spherical curvature does not directly correspond to the OLED location, then no one can further tune the far field pattern due to refraction effects since the light rays will not be traveling
 5 through the lens/air interface (or the shaped substrate/air interface) at normal incidents. Other methods which break the planarity of the back surface of the substrate will also achieve less total internal reflection, and bus increased external efficiency.

Applicants show below a plurality of embodiments for the invention
 10 which they have implemented and developed experimental results. Various parameters material to the implementation of those embodiments are summarized in Table 1 hereafter. Three derived parameters shown in Table 1 can best be understood that with reference to Figure 2. The first of these derived parameters is the substrate index (n_{subs}), which completely determines $\theta_{org, c1}$ and $\theta_{org, c2}$ and
 15 therefore also completely determines η_{ext} and η_{subs} . n_{subs} also determines the intensity distribution profile inside the substrate layer, which can be obtained from Equation (3) by replacing n_{air} with n_{subs} [See, G. Gu, *et al.*, *id.*]

$$I_{subs}(\theta_{subs}) = \frac{F}{2\pi} \frac{n_{subs}^2 \cos \theta_{subs}}{n_{org}^2 \sqrt{1 - \left(\frac{n_{subs}}{n_{org}} \sin \theta_{subs} \right)^2}} \quad (4)$$

$I_{subs}(\theta_{subs})$ is of interest because Equation (3) only describes the external intensity
 20 distribution when the substrate is planar. If the substrate forms a hemisphere with the device at its center, for instance, $I_{ext}(\theta_{ff})$ will be equal to $I_{subs}(\theta_{subs})$. In fact, $I_{subs}(\theta_{subs})$ plays a direct role in determining $I_{ext}(\theta_{ff})$ in all cases *except* the special case of a planar substrate. Continuing, the effect of n_{subs} on I_{subs} is to focus the distribution as n_{subs} is increased, until $n_{subs}=n_{org}$, at which point $I_{subs}(\theta_{subs})$
 25 reproduces the isotropic intensity distribution initially generated in the organic layer.

The second of the derived parameters is the maximum angle of light (in the substrate) collected into the lens ($\theta_{subs, max}$). This parameter is obtained from the total substrate thickness (t_{subs}) and the lens radius (ρ_{lens}) by the expression,

$\theta_{\text{subs, max}} = \tan^{-1}(\rho_{\text{lens}}/t_{\text{subs}})$. Assuming $\theta_{\text{subs, max}}$ is large enough to capture all of the light that would have been transmitted out of the substrate without the lens (i.e. $\theta_{\text{subs, max}} > \sin^{-1}(n_{\text{air}}/n_{\text{subs}})$), then any light emitted at angles greater than $\theta_{\text{subs, max}}$ will be wave-guided in the substrate and lost. Following the same analysis which
 5 led to Equation (1), an expression can now be obtained under the same assumption on $\theta_{\text{subs, max}}$ for the external coupling efficiency for an OLED centered over a spherically shaped substrate,

$$\eta_{\text{ext}} = \int_0^{\theta_{\text{subs, max}}} I_{\text{subs}}(\theta_{\text{subs}}) \sin(\theta_{\text{subs}}) d\theta_{\text{subs}} \quad (\text{spherical substrate}). \quad (5)$$

To demonstrate the significance of $\theta_{\text{subs, max}}$, consider that if $\theta_{\text{subs, max}} = 76^\circ$ (i.e.
 10 $\rho_{\text{lens}} = 4 t_{\text{subs}}$) and $n_{\text{subs}} = n_{\text{org}}$, that uncoupled 14° represents 24 percent of the light transmitted into the substrate.

The third and final of the derived parameters is the vertical offset of the device from the center of curvature of the lens (d_{offset}). This parameter is of interest because it strongly affects the far field distribution pattern. The analytical
 15 expression for I_{ext} when $d_{\text{offset}} \neq 0$ can readily be found in the art. For purposes of this discussion, it is sufficient to point out that when the OLED is positioned too far from the lens ($d_{\text{offset}} > 0$), I_{ext} will be more focused than I_{subs} , and when the OLED is positioned too closely to the lens ($d_{\text{offset}} < 0$), I_{ext} will be less focused than I_{subs} . However, over a wide range of offset values, there is only a minor
 20 degrading effect on η_{ext} due to $|d_{\text{offset}}| > 0$.

Note that the ray used to define the far-field angle, θ_{ff} , of Figure 2(a), is drawn for the $d=0$ case, while in the diagram, d , the offset between the center of curvature of the lens and the OLED, is drawn as non-zero so that it can be clearly identified. Note also that spherical features implemented as a plastic lens array
 25 laminated to a planar substrate are particularly illustrated in Figure 2(b).

While it is noted that the basic substrate-face design has been used before with traditional LEDs, that approach has not previously been applied for the improvement of output coupling in OLEDs. In addition, such an OLED approach would not have been suggested by the LED application. Note as well that the
 30 extremely high indices of refraction (e.g. $n > 4$) found in the emitting materials in

traditional LEDs precluded some of the advantages one can obtain with OLEDs. In particular, by matching the index of the substrate to the index of the emitting material in addition to shaping the substrate, one can potentially eliminate *all* of the external coupling losses in the device, and transparent substrate materials in the appropriate index range (*i.e.* $n \sim 1.6 - 1.8$) are readily available.

The inventors have implemented the approach of the invention for a number of embodiments, which are further described hereafter. The OLEDs that constitute these various embodiments were fabricated on glass and polycarbonate (PC) substrates. The glass substrate consisted of 0.7 mm and 1.1 mm-thick soda lime glass purchased from Applied Films Co. coated with ITO by the manufacturer. The PC substrates consisted of 175 μm -thick sheets purchased from Goodfellows Co., with a 100 nm ITO film deposited on it in an Edwards A306 RF magnetron sputter with 2 mTorr pure Ar gas at 150 W RF power at room temperature. The sputter target was 90% In_2O_3 -10% SnO, 3 in. in diameter. The deposition rate was 33 nm/min. The OLEDs were made by spinning on a single poly-(N-vinylcarbazole) (PVK)/2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole(PBD)/Coumarin 6 (C6) layer, and evaporating a 100 to 200 nm Mg:Ag cathode [Wu]. The index of refraction of the organic layer was measured to be 1.67 by ellipsometry at $\lambda = 634$ nm and $\lambda = 830$ nm. Typical device size and geometry consisted of a circle 1.75 mm in diameter.

Experiments were performed with six different substrate structures, which are designated Trials 1 through 6 in Table 1. A diagram of the substrate setup (with all relevant parameters identified) is provided in Figure 2(a).

Table 1

Trial	Substrate Material	Lens Material	R_{lens} (mm)	ρ_{lens} (mm)	L_{cath} (mm)	θ_{max}	d (mm)	$I_{\text{normal}}/I_0 \pm 0.1$	$F/F_0 \pm 0.1$
1	Glass ($n=1.51$)	N/A	N/A	N/A	0.7	N/A	N/A	1.0	1.0
2	Glass ($n=1.51$)	Glass ($n=1.51$)	3.4	3.4	0.7	78°	+1.0	3.6	2.0
3	Glass ($n=1.51$)	Glass ($n=1.51$)	3.4	3.4	2.0	60°	+2.3	9.5	1.6
4	Glass ($n=1.51$)	Silicone ($n=1.41$)	2.7	2.4	1.9	51°	+0.6	2.1	1.6
5	PC ($n=1.61$)	N/A	N/A	N/A	1.0	N/A	N/A	1.0	1.0
6	PC ($n=1.61$)	Epoxy ($n=1.60$)	2.7	2.4	1.0	67°	-0.3	1.6	3.0

Substrate and lens parameters (as defined in Figure 2a) for different external coupling experiments. I_{normal}/I_0 and F/F_0 represent the ratio of normal emission intensity and total emission intensity respectively to the results obtained for identical devices fabricated on planar substrates of

the same substrate material. The total emission intensity measurement does not include edge emission.

In Trial 1, unmodified planar glass substrates were used. In Trials 2 and 3,
5 glass substrates with attached glass condenser lenses purchased from Edmund
Scientific Co. were used. In Trial 4, glass substrates with attached molded
silicone lenses were used. In these latter three trials, the lenses were attached to
the substrate using index matching gel purchased from FIS Co.. When necessary,
the substrate thickness was increased by gluing on extra glass slides with the same
10 index matching gel. The silicone lenses were molded in a Teflon block with ball-
milled indentations with GE RTV615 silicone ($n = 1.405$). In Trials 5 and 6, PC
substrates were used. In Trial 5, the standard planar substrate was employed,
while in Trial 6, molded epoxy lenses were attached to the PC substrate. The
epoxy lenses were fabricated using the same mold as described above with Master
15 Bond EP42HT two-component epoxy ($n = 1.61$). The lenses were glued to the
substrate with uncured epoxy. For each trial, the far field intensity pattern was
measured using a large area (1 cm diameter) Si photodetector at a distance of 10
cm away from the device at 6° increments between 0° (normal) and 90° .

The three derived parameters previous described and shown in Figure 2(a)
20 -- substrate index (n_{subs}), maximum angle of light collected into the lens ($\theta_{\text{subs, max}}$), and vertical offset of the device from the center of curvature of the lens (d_{offset}) -- all play a critical role in determining the total emitted flux and the external intensity distribution in that experiment. With these parameters in hand, it is a simple matter to evaluate the experimental results. The normal and total
25 emitted flux measured in each trial is listed in Table 1. Plots of the measured emitted flux at each angle for each trial are provided in Figures 3b and 3c. The total emitted flux results are initially addressed.

In the trials with glass substrates (Trials 2 – 4), the total emitted flux was increased by a factor of between 1.6 and 2.0 ± 0.1 . These results are consistent
30 with the analytical values obtained from Equations (5) and (1), where it is noted that in Trial 3, the large d_{offset} is expected to slightly degrade the effectiveness of the lens. (Because $\theta_{\text{subs, max}} < \sin^{-1}(n_{\text{silicone}}/n_{\text{subs}})$, the lower index of the silicone does not have a significant effect on the results.) In the trial with the PC substrate

and an epoxy lens (Trial 6), the total emitted flux was increased by a factor of 3.0 ± 0.1 over the planar PC substrate case (Trial 5), which is again consistent with the analytical values obtained using Equations (5) and (1). It is noted that Equation (1) predicts that the planar cases for the PC and glass substrates should be identical, it can be observed in Figure 3c that the two cases (Trials 1 and 5) are indeed effectively identical within experimental uncertainties of the trials. This provides experimental justification for comparing the results obtained for PC substrates with those for glass substrates.

The emitted flux distribution results are now addressed. In all of the results, one can correlate the normal emitted flux with the value for d_{offset} . Trial 3 represents one extreme: $d_{\text{offset}} = 2.3$ mm and $I_{\text{normal}}/I_0 = 9.5$. Trial 6 represents the other extreme: $d_{\text{offset}} = -0.3$ mm and $I_{\text{normal}}/I_0 = 1.6$. Since different substrate and lens materials were used in each trial, the value of I_{normal}/I_0 is not *solely* determined by d_{offset} ; however, the analytical expression for I_{ext} and our results demonstrate that for the normal 6° cone, d_{offset} is the dominant parameter. In Figures 3b and 3c, the complete emitted intensity distribution results are presented. These results again demonstrate the focusing effect of increasing d_{offset} . In addition, the results from Trial 6 demonstrate just how effective using a lens with a high index substrate can be at reproducing the isotropic intensity distribution generated in the organic layer -- the emitted flux remains effectively constant from 0° to 72° in this trial.

It is noted finally that, in Figures 3(a) and 3(b), one sees essentially perfect correlation between the data for the planar glass substrate (Trial 1) and the two different refraction models described above. Furthermore, the data significantly deviates from the Lambertian distribution results, further reinforcing the validity of the refraction models presented in this paper.

Having thus described the invention in detail, it is to be understood that the foregoing description is not intended to limit the spirit and scope thereof. What is desired to be protected by Letters Patent is set forth in the appended claims.

CLAIMSWhat is claimed is:

1. A light emitting device, said device comprising:
 - 5 a transparent substrate having a first surface and a second surface;
 - a conductive transparent layer disposed on said first surface of said substrate;
 - an organic layer disposed on said conductive oxide layer; and
 - a top contact disposed on said organic layer;
- 10 wherein a contour of said second surface of said substrate is caused to assume a non planar form.
2. The light emitting device of claim 1 wherein said non-planar form is spherical.
3. The light emitting device of claim 1 wherein said second surface contour is caused to assume a non-planar form by moulding said substrate surface.
4. The light emitting device of claim 1 wherein said second surface contour is caused to assume a non-planar form by laminating a first surface of a second transparent layer having a non-planar second surface to said second surface of said substrate.
5. A method for increasing light emissivity for organic light emitting diodes (OLED), wherein said OLED is disposed on a first surface of a transparent substrate, said method comprising the step of:
 - 15 causing a second surface of said substrate to assume a non-planar form.
6. The method for increasing light emissivity for OLEDs of claim 5 wherein said non-planar form is spherical.
7. The method for increasing light emissivity for OLEDs of claim 5 wherein said second surface contour is caused to assume a non-planar form by moulding said substrate surface.
8. The method for increasing light emissivity for OLEDs of claim 5 wherein said second surface contour is caused to assume a non-planar form by laminating a first surface of a second transparent layer having a non-planar second surface to said second surface of said substrate.

9. A method for constructing a light emitting device comprising the steps of:
providing a transparent substrate having a first and a second surface;
depositing a conductive transparent layer on said first surface of said
substrate;
- 5 depositing an OLED layer on said conductive transparent layer; and
causing a contour of said second surface of said substrate to assume a non-
planer form.
10. The method for constructing a light emitting device of claim 9 wherein
said non-planar form is spherical.
11. The method for constructing a light emitting device of claim 9 wherein
said second surface contour is caused to assume a non-planar form by moulding
said substrate surface.
12. The method for constructing a light emitting device of claim 9 wherein
said second surface contour is caused to assume a non-planar form by laminating
a first surface of a second transparent layer having a non-planar second surface to
said second surface of said substrate.
13. A light emitting device, said device comprising:
a transparent substrate having a first surface and a second surface;
an OLED layer disposed on said first surface of said substrate;
wherein said second surface of said substrate is molded to a non-planar
form.
14. The light emitting device of claim 13 wherein said non-planar form is
spherical.
15. A light emitting device, said device comprising:
a transparent substrate having a first surface and a second surface;
10 an OLED layer disposed on said first surface of said substrate;
wherein a contour of said second surface is caused to assume a non-planar
form by laminating a first surface of a second transparent layer having a non-
planar second surface to said second surface of said substrate.
16. The light emitting device of claim 15 wherein said non-planar form is
spherical.

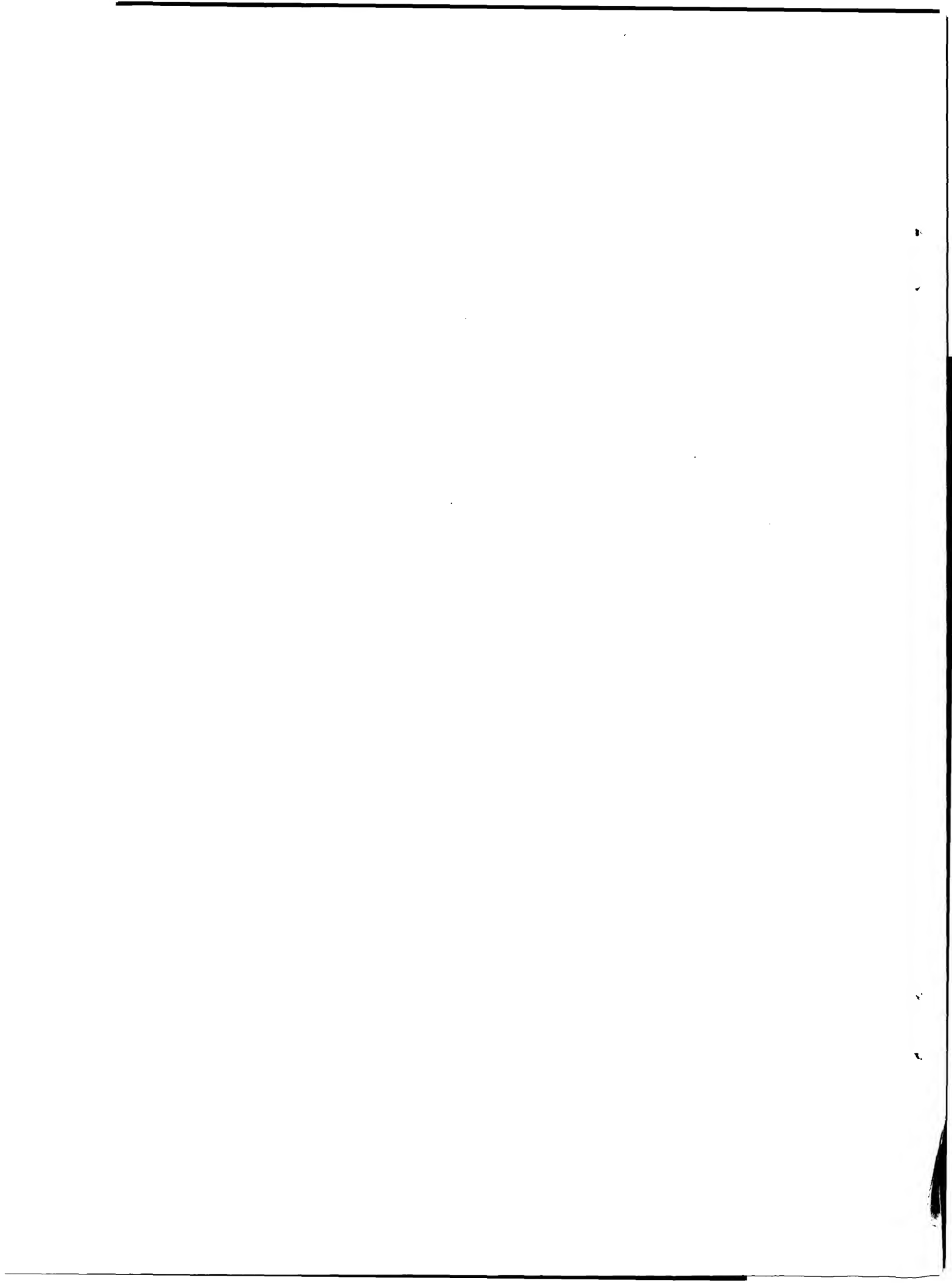
17. A method for increasing light emissivity of a light emitting device, wherein said light emitting device includes at least two OLEDs disposed on a first surface of a transparent substrate, said method comprising the step of:

causing a contour of selected portions of a second surface of said substrate
5 to assume a non-planar form, said selected portions being selected to be in optical alignment with a light output from ones of said at least two OLEDs.

18. The method for increasing light emissivity of claim 17 wherein wherein said non-planar form is spherical.

19. The method for increasing light emissivity of claim 17 wherein said selected second surface contours are caused to assume a non-planar form by moulding said substrate surface.

20. The method for increasing light emissivity of claim 17 wherein said selected second surface contours are caused to assume a non-planar form by laminating a first surface of a second transparent layer having a non-planar second surface to said second surface of said substrate.



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FIG. 1

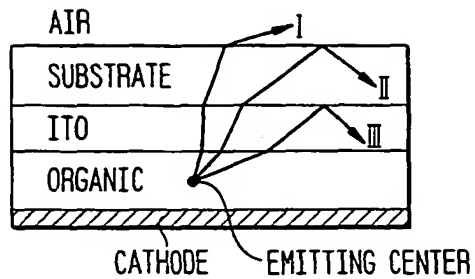


FIG. 2A

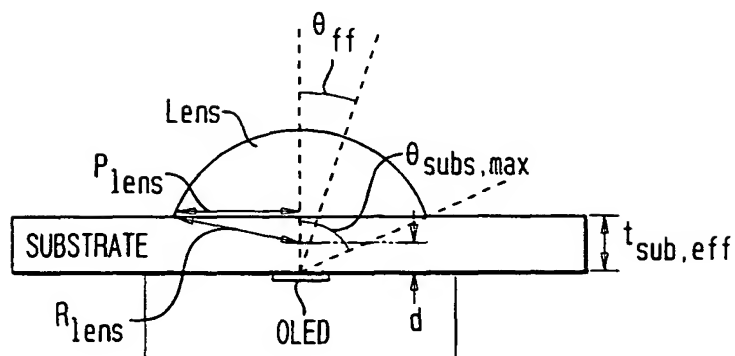
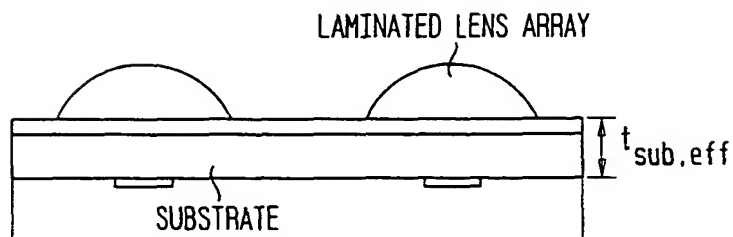


FIG. 2B



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FIG. 3A

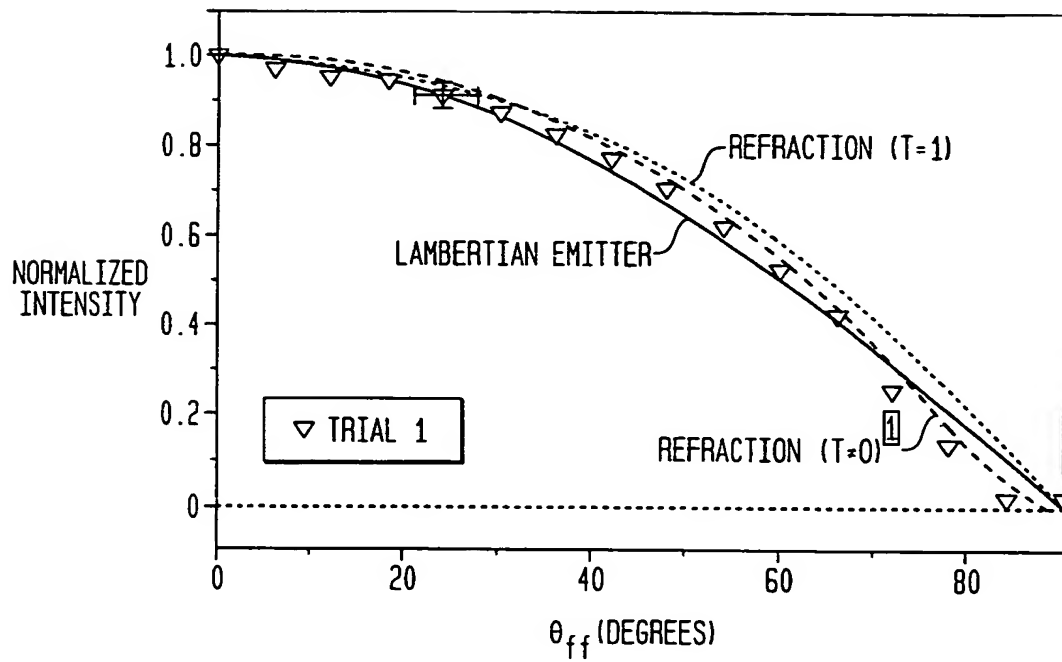
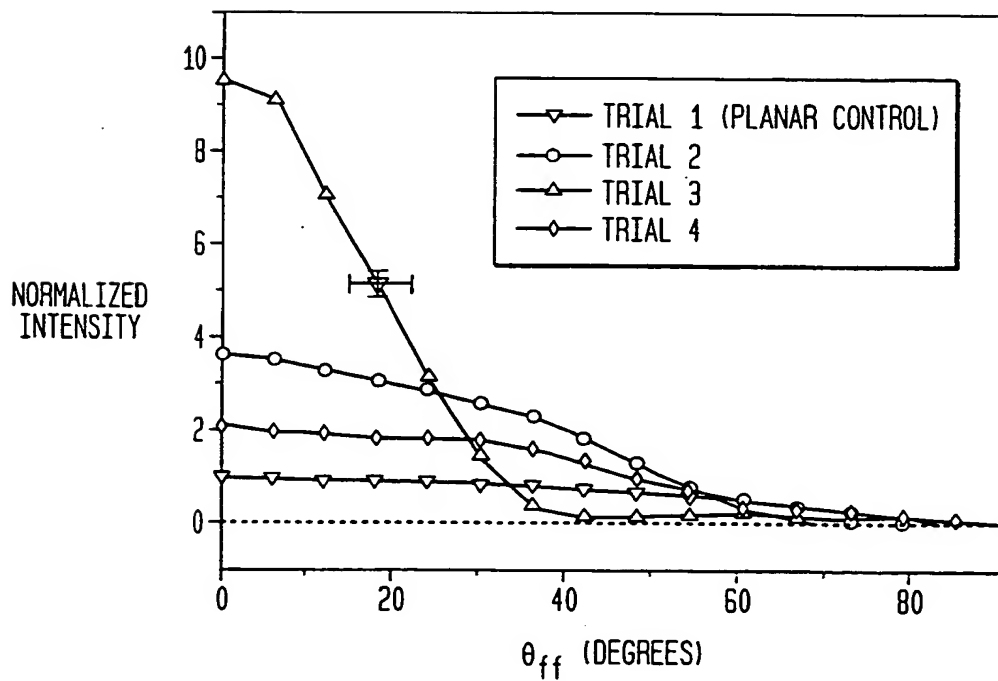


FIG. 3B





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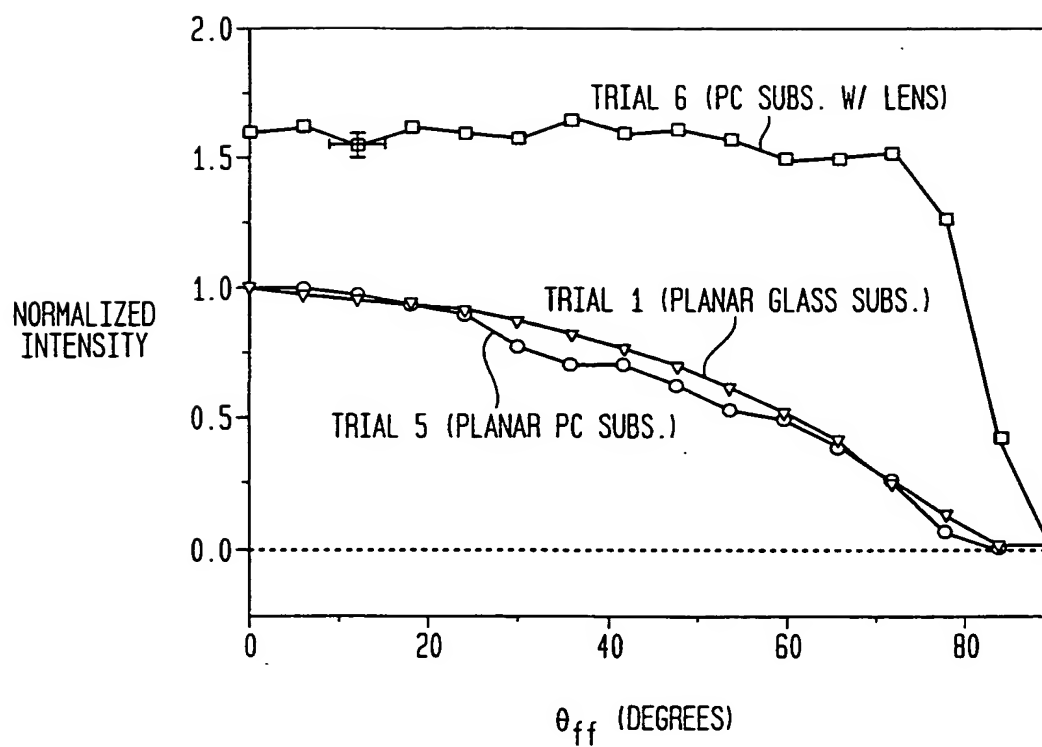
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FIG. 3C





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INTERNATIONAL SEARCH REPORT

International application No.

PCT/US00/29822

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : H01J 1/62, 63/04

US CL : 313/495,496,497,498,499,500,501,502,503,504,505,506,507,508,509,512; 428/690,917; 445/24

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 313/495,496,497,498,499,500,501,502,503,504,505,506,507,508,509,512; 428/690,917; 445/24

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EAST

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5,371,434 A (RAWLINGS) 06 December 1994 (06.12.1994)	1-20
A	US 5,618,626 A (NAGASHIMA et al.) 08 April 1997 (08.04.1997)	1-20
A	US 5,814,416 A (DODABALAPUR et al.) 29 September 1998 (29.09.1998)	1-20
A	US 5,936,347 A (ISAKA et al.) 10 August 1999 (10.08.1999)	1-20

☐ Further documents are listed in the continuation of Box C.

☐ See patent family annex.

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document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents; such combination being obvious to a person skilled in the art

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16 January 2001 (16.01.2001)

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